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# Research Article

# Thermophysical Properties of 1-Octyl-3-Methylimidazolium Acetate with Organic Solvents

## Fuxin Yang and Peng Feng

Key Laboratory of Thermo-Fluid Science and Engineering, Ministry of Education, Xi'an Jiaotong University, Xi'an, Shaanxi 710049, China

Correspondence should be addressed to Fuxin Yang; fxyang@xjtu.edu.cn

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Ionic liquid (IL) usually possesses high viscosity. In this work, the selected organic solvents, namely, dimethyl sulfoxide, N,N-dimethylacetamide, and N,N-dimethylformamide, were used as the diluents to lower IL viscosity. The thermophysical properties of the densities and viscosities for the binary mixtures of IL (i.e., 1-octyl-3-methylimidazolium acetate) with solvents were studied at normal pressure in the temperature ranges of 303.15 K to 348.15 K. The effects of the organic solvents on lowering IL viscosity were quantitatively evaluated. The excess properties of the mixtures were calculated to analyze the interactions between IL and solvents. The hard-sphere model was employed to reproduce the viscosity behavior of the pure substances and binary mixtures.

#### 1. Introduction

Ionic liquids (ILs) are considered as the novel class of molten salts. They consist of the organic cations and organic anions or inorganic anions. The different anion and cation can be combined and synthetized as the new ionic liquid to meet the requirements for the application. Because of the tunable characteristics, there are huge ionic liquids to be explored. Moreover, due to their promising properties, such as the thermal stability, the good conducting nature, and the negligible vapor pressure, ILs are recognized as the green and environmentally friendly solvents, receiving extensive studies [1–3].

The imidazolium-based ILs, such as 1-alkyl-3-methylimidazolium chloride and 1-alkyl-3-methylimidazolium acetate, have been studied in the biofuel process and shown the ability to dissolve the lignocellulosic biomass [4]. Yang et al. and Wang et al. studied the oxidation cleavage of  $\beta$ -O-4 bond in lignin model compounds and lignin dissolution using the IL of 1-octyl-3-methylimidazolium acetate ([C<sub>8</sub>mim][OAc]) [5, 6]. Zhang et al. reported the effect of the cation and anion structure of ionic liquids on the solubility of cellulose; however, [C<sub>8</sub>mim][OAc] exhibited poor

dissolving capability due to its high viscosity that impedes the cellulose dissolution in ILs [7]. One of the possible ways to reduce the effects of IL high viscosity is to use solvent as the diluent or additive into IL. Dimethyl sulfoxide (DMSO) is considered as the alternative solvent and the IL-DMSO mixture has been conducted in the biofuel production and biomass depolymerization [8]. The aprotic solvents of N,N-dimethylacetamide (DMA) and N,N-dimethylformamide (DMF) are also well studied [9]. Furthermore, publications have been focused on the influences of organic solvents on the properties of the binary mixtures containing IL with solvents [10, 11]. The thermophysical properties, namely, the density and the viscosity, are the fundamental data for the scientific research and the industrial application, especially for industrial process design.

In this work, the viscosities and densities of 1-octyl-3-methylimidazolium acetate together with the three organic solvents of DMSO, DMA, and DMF were conducted at atmospheric pressure (0.0967  $\pm$  0.0020) MPa from 303.15 K to 348.15 K. The properties of [C<sub>8</sub>mim][OAc] with DMF were not reported in the published studies as well as the binary mixture of [C<sub>8</sub>mim][OAc] with DMSO and DMF in IL mole fraction from 0.1 to 1. The viscosity and density of

the pure substances and IL with solvents were measured using a capillary viscometer and an Anton-Paar vibrating U-tube densimeter, respectively. The effects of solvents on IL thermophysical properties were quantitatively studied. The excess properties of the mixtures were calculated for analyzing the interactions between ionic liquid with the solvents. The hard-sphere model was employed to reproduce the viscosity.

#### 2. Materials and Methods

2.1. Materials. IL of 1-octyl-3-methylimidazolium acetate was supplied by Shanghai Weiting Reagent Biological Technology Co., Ltd. The organic solvents were obtained from Aladdin Shanghai Biochemical Technology Co., Ltd. The solvents were used in the experiment measurement without further treatment. IL was treated using the molecular sieves (3A, 1.6 mm pellets) obtained from Sigma-Aldrich (St Louis, MO). More details about the solvents and IL are summarized in Table 1. The chemical structure and physical properties of IL are listed in Table S1 (in the Supplementary Material).

2.2. Sample Treatment. The water content in ionic liquid is considered as the impurity and it would obviously affect the thermophysical properties of IL, especially of viscosity. To eliminate the influences, water was extracted from IL using the molecular sieves. The sieves were rinsed by acetone and methanol. The clean sieves were then put into an oven at 473.15 K for more than 8 hours. IL was mixed with the treated molecular sieves and they were placed in a vacuum at 353.15 K with the pressure of less than 2 kPa for 48 hours. The mixtures of ionic liquid with solvents were weighed using a balance (ME204, Mettler Toledo) and its uncertainty was 0.0001 g. The water content in the sample was detected by a moisture titrator (Kyoto Electronics Manufacturing Co., Ltd.) with less than 0.3 wt. %.

2.3. Thermophysical Properties Measurement. The density was measured using an Anton-Paar vibrating U-tube densimeter (5000 M). The hot water and methanol were used to clean the densimeter. The uncertainty of temperature was 0.01 K. The relative standard uncertainty of density was 0.005. The density data was measured three times.

The viscosity was measured using an Ubbelohde viscometer (9721-R53, 9721-R56, 9721-R59, 9721-R65, and 9721-R71) supplied by Cannon Instrument Company (Pennsylvania, USA). An immersion thermostat (LAUDA ECO Silver) was used to control an oil bath for the capillary cell and the temperature uncertainty was 0.01 K. The flow time was determined by a stopwatch. The dynamic viscosity  $\eta$  was calculated as follows:

$$\eta = kt\rho,\tag{1}$$

where k is the viscometer constant and it is determined using the standard solution, t is the efflux time of the sample, and  $\rho$  is the density corresponding to the same measurement condition.

The efflux time of the sample was more than 100 s and, the diameter of the capillary was 0.47 mm that was less than the length of 290 mm; therefore, in the experiment, the kinetic energy was ignored as well as the end correction. Moreover, because the value of IL viscosity is always large, the Ubbelohde capillary with large viscometer constant was used for IL measurement (9721-R65 and 9721-R71), while the Ubbelohde capillary with small viscometer constant was performed for the organic solvents (9721-R53). The relative combined standard uncertainty of the viscosity in the measurement was 0.06. The viscosities were measured three times.

In Table 2, the experimental densities and viscosities data in this work as well as the literature values at atmospheric pressure are presented to check the apparatus and the chemicals [12–21]. It is found that the densities and viscosities agree well with the literature values.

#### 3. Results and Discussion

3.1. Experimental Density Data. In general, the experimental density data can be fitted as a linear equation:

$$\rho = a + b \cdot T,\tag{2}$$

where  $\rho$  is the density data in kg·m<sup>-3</sup>, the fitted parameters of a (kg·m<sup>-3</sup>) and b (kg·m<sup>-3</sup> K<sup>-1</sup>) are correlated using the experimental density, and T (K) is the measurement temperature.

To check the deviation between the literature data and the experimental value, the average absolute relative deviation (AARD) is calculated as follows:

$$AARD(\%) = \frac{100}{n} \sum_{i=1}^{n} \left| \frac{E_{(exp/lit),i} - C_{cal,i}}{E_{(exp/lit),i}} \right|, \tag{3}$$

where n is the data points,  $E_{\text{exp/lit,i}}$  is the experiment/literature value, and  $C_{\text{cal,i}}$  represents the calculated data.

Table S2 gives the parameters for (2). The calculated density data of the three solvents are compared with those in the literature, as shown in Figures S1–S3 (in the Supplementary Material). The AARD is 0.04% for DMSO [12, 14, 22–26], 0.28% for DMA [27–34], and 0.05% for DMF [14, 20, 27, 28, 35–38]. The deviations between the literature data and the experiment values in this study are small. Therefore, the density data for the mixtures of ionic liquid with solvents are measured and summarized in Tables 3–5.

Figure 1 and Figures S4–S6 give the density as a function of temperature. It is found that the data increase with the decrease of the temperature. Generally, the temperature will affect the kinetic energy of the atom and the molecule. When the temperature increases, the kinetic energy increases as well as the volume of the substance; then the density decreases accordingly. In Figure 1, the values of densities can be ordered as follows: DMSO > IL > DMF > DMA. Among these chemicals, the curve of DMSO is the sharpest. IL presents the less sharp curve and the absolute value of IL slope is the smallest.

In the density measurement of binary mixture, the excess molar volume is introduced to study the consistency of the

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TABLE I: The	descriptions	of ionic	liquid and	l organic solvents.

IL/solvent	Abbreviation	Initial mass fraction purity	Treatment	Source	Cas no.
1-Octyl-3-	[C <sub>8</sub> mim]	≥0.98	Dry using	Shanghai Weiting Reagent Biological	366491-
methylimidazolium acetate	[OAc]		molecular sieve	Technology Co., Ltd.	21-6
Dimethyl sulfoxide	DMSO	≥0.98	None	Aladdin Shanghai Biochemical Technology Co., Ltd.	67-68-5
N,N-dimethylacetamide	DMA	≥0.98	None	_	127-19-5
N,N-dimethylformamide	DMF	≥0.98	None	_	68-12-2

Table 2: Densities and viscosities of the pure chemicals at normal pressure (0.0967 ± 0.0020) MPa\*.

IL/solvent		Density/kg	$\cdot \text{m}^{-3}$	Viscosity (mPa·s)		
IL/Solveilt	Temperature (K)	This work	Literature	This work	Literature	
DMSO	303.15	1090.4	1090.73 [12], 1090.41 [13]	1.81	1.786 [14], 1.84 [15], 1.961 [16]	
DMA	303.15	932.0	931.66 [17], 931.69 [13]	0.88	0.8769 [18], 0.87 [19]	
DMF	303.15	939.7	939.34 [17], 939.23 [13]	0.78	0.766 [20], 0.7647 [21]	

<sup>\*</sup>The standard uncertainty (u) is u(T) = 0.01 K. The relative standard uncertainty ( $u_r$ ) is  $u_r(\rho) = 0.005$ . The relative combined standard uncertainty ( $u_{c,r}$ ) is  $u_{c,r}(\eta) = 0.06$ .

Table 3: Experimental densities of [C8mim][OAc] with organic solvent of DMSO at atmospheric pressure (0.0967  $\pm$  0.0020) MPa from 303.15 K to 348.15 K.

-				<i>x</i> [C <sub>8</sub> mim][0	$OAc] + (1-x) \Gamma$	MSO, ρ/kg·m	-3		
T/K					x				
	1.000	0.842	0.735	0.553	0.415	0.312	0.234	0.172	0.000
303.15	988.8	994.5	999.6	1010.0	1020.8	1031.1	1041.0	1050.7	1090.4
308.15	985.9	991.5	996.5	1006.7	1017.4	1027.5	1037.3	1046.7	1085.5
313.15	982.9	988.4	993.3	1003.5	1014.0	1023.9	1033.5	1042.7	1080.5
318.15	979.9	985.4	990.2	1000.2	1010.6	1020.4	1029.7	1038.8	1075.5
323.15	976.9	982.3	987.1	997.0	1007.2	1016.8	1025.9	1034.8	1070.5
328.15	973.9	979.3	984.0	993.8	1003.8	1013.2	1022.1	1030.8	1065.5
333.15	970.9	976.2	980.9	990.5	1000.4	1009.6	1018.3	1026.8	1060.5
338.15	967.9	973.1	977.8	987.3	997.0	1006.0	1014.5	1022.8	1055.5
343.15	964.9	970.1	974.6	984.0	993.6	1002.4	1010.7	1018.8	1050.5
348.15	961.9	967.1	971.5	980.8	990.2	998.8	1006.9	1014.7	1045.5

The standard uncertainties (u) are  $u(x) = 2.0 \cdot 10^{-3}$  and u(T) = 0.01 K. The relative standard uncertainty ( $u_r$ ) is  $u_r(\rho) = 0.005$ .

Table 4: Experimental densities of [C8mim][OAc] with organic solvent of DMA at atmospheric pressure (0.0967  $\pm$  0.0020) MPa from 303.15 K to 348.15 K.

				$x [C_8 mim][O$	Ac] + (1-x) D	MA, $\rho/\text{kg}\cdot\text{m}^{-3}$			
T/K					x				
	1.000	0.861	0.751	0.581	0.445	0.343	0.257	0.187	0.000
303.15	988.8	986.1	983.7	978.9	973.3	967.8	961.9	955.8	932.0
308.15	985.9	983.1	980.5	975.6	969.9	964.2	958.1	951.8	927.4
313.15	982.9	980.0	977.4	972.3	966.4	960.5	954.3	947.8	922.8
318.15	979.9	976.9	974.2	969.0	963.0	956.9	950.5	943.8	918.1
323.15	976.9	973.8	971.1	965.7	959.5	953.3	946.7	939.8	913.5
328.15	973.9	970.8	967.9	962.4	956.0	949.6	942.9	935.8	908.9
333.15	970.9	967.7	964.8	959.1	952.5	946.0	939.1	931.5	904.2
338.15	967.9	964.6	961.6	955.8	949.0	942.3	935.2	927.8	899.6
343.15	964.9	961.6	958.5	952.5	945.5	938.7	931.4	923.8	894.9
348.15	961.9	958.5	955.3	949.2	942.0	935.0	927.5	919.7	890.2

The relative standard uncertainty  $(u_r)$  is  $u_r(\rho) = 0.005$ .

				$x [C_8 mim][O$	[Ac] + (1-x) D	MF, $\rho/\text{kg}\cdot\text{m}^{-3}$			
T/K					x				
	1.000	0.842	0.723	0.530	0.407	0.299	0.224	0.160	0.000
303.15	988.8	986.7	984.7	980.1	975.8	970.8	966.1	960.6	939.7
308.15	985.9	983.6	981.6	976.8	972.3	967.1	962.3	956.6	934.9
313.15	982.9	980.5	978.4	973.4	968.8	963.4	958.4	952.6	930.1
318.15	979.9	977.5	975.3	970.1	965.3	959.7	954.6	948.5	925.3
323.15	976.9	974.4	972.1	966.7	961.8	956.0	950.7	944.4	920.5
328.15	973.9	971.3	968.9	963.3	958.3	952.3	946.8	940.4	915.7
333.15	970.9	968.2	965.7	960.0	954.8	948.6	942.9	936.3	910.9
338.15	967.9	965.2	962.6	956.6	951.2	944.9	939.0	932.2	906.1
343.15	964.9	962.1	959.4	953.2	947.7	941.1	935.0	928.1	901.2
348.15	961.9	959.0	956.2	949 8	944 1	937 4	931.0	924 0	896 3

Table 5: Experimental densities of [C8mim][OAc] with organic solvent of DMF at atmospheric pressure (0.0967  $\pm$  0.0020) MPa from 303.15 K to 348.15 K.

The relative standard uncertainty  $(u_r)$  is  $u_r(\rho) = 0.005$ .

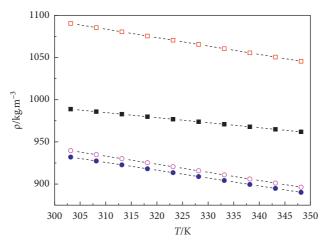


FIGURE 1: Density of [C8mim][OAc] and organic solvents as a function of temperature at normal pressure. ■: IL; □: DMSO; ●: DMA; O: DMF. Dash line calculated by (2).

experiment density data [39]. The excess molar volume  $V^{E}$  is obtained by [40]

$$V^{E} = V_{m} - \sum x_{i} V_{i} = \frac{\sum x_{i} M_{i}}{\rho_{m}} - \sum \frac{x_{i} M_{i}}{\rho_{i}}$$
$$= \sum x_{i} M_{i} \left(\frac{1}{\rho_{m}} - \frac{1}{\rho_{i}}\right), \tag{4}$$

where  $V^{\rm E}$  is excess molar volume in m<sup>3</sup>·mol<sup>-1</sup>. The subscripts of "m" and "i" mean the properties of mixture and pure substance i, respectively. X is IL mole fraction. V (m<sup>3</sup>·mol<sup>-1</sup>),  $\rho$  (kg·m<sup>-3</sup>), and M (kg·mol<sup>-1</sup>) are the molar volume, density, and the molar mass, respectively. The combined standard uncertainty of excess molar volume is less than  $2.24 \times 10^{-7}$  m<sup>3</sup> mol<sup>-1</sup>.

The Redlich-Kister equation is introduced to fit excess molar volume [41, 42]:

$$V^{E} = x(1-x)\sum_{i=0}^{n} A_{i}(2x-1)^{i},$$
 (5)

where x is IL mole fraction and  $A_i$  (m<sup>3</sup>·mol<sup>-1</sup>) is the fit parameter correlated by experiment data in this work. Table S3 gives the fit parameters and AARD.

Figures 2–5 depict  $V^{\rm E}$  as a function of IL mole fraction. The  $V^{\rm E}$  values are negative, indicating that the volumes of mixtures become smaller than the ideal ones and there should be interactions between ionic liquid with solvents. In the previous studies, the negative  $V^{\rm E}$  values were also observed for the mixture of 1-alkyl-3-methylimidazolium acetate with organic solvents, and the excess volume becomes more negative when the temperature increases [43, 44]. The curves are asymmetric and the minimum values are detected in dilute regions.

Furthermore, Apelblat proposed a new two-parameter equation for correlation and prediction of densities as a

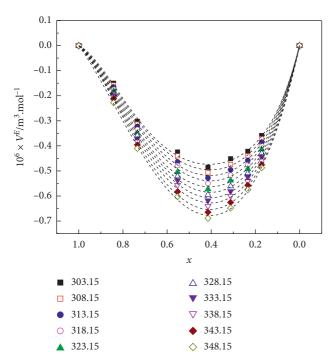


FIGURE 2: The excess molar volume of [C8mim][OAc] with solvent of DMSO as a function of IL mole fraction at atmospheric pressure; dash line calculated by (5).

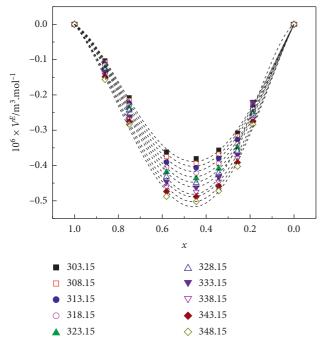


FIGURE 3: The excess molar volume of [C8mim][OAc] with solvent of DMA as a function of IL mole fraction at atmospheric pressure; dash line calculated by (5).

function of temperature and concentration in binary aqueous solutions [45]. The equation is as follows:

$$F(x) = \frac{1}{\rho_{\rm IL}(T)} - \frac{1}{\rho_m(T, x)}.$$
 (6)

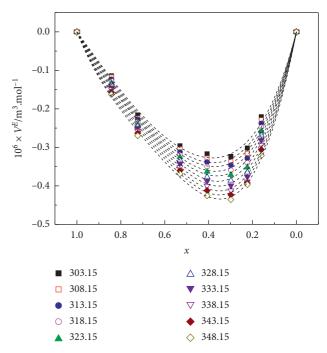


FIGURE 4: The excess molar volume of [C8mim][OAc] with solvent of DMF as a function of IL mole fraction at atmospheric pressure; dash line calculated by (5).

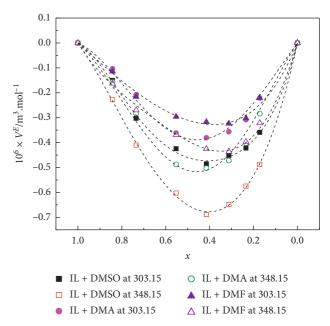


FIGURE 5: The excess molar volumes of [C8mim][OAc] with solvents as a function of IL mole fraction; dash line calculated by (5).

An expression of  $F(x) = Ax + Bx^2$  was suggested; however, the errors were nonnegligible for the binary mixtures containing IL. In this work, a modified form was used:

$$F(x) = C_0 + C_1 x + C_2 x^2 + C_3 x^3, (7)$$

where  $\rho_{\rm IL}$  (*T*),  $\rho_{\rm m}$  (*T,x*) are the densities of IL and binary mixture, respectively. *X* is the IL mole fraction. The

(1-x) solvent	x IL	η₀/mPa·s	B/K	$T_0/K$	AARD/%	$E_{\eta (303.15 \text{ K})}/\text{kJ}\cdot\text{mol}^{-1}$
	1	0.0575	1129.78	171.38	0.46	49.72
	0.842	0.0033	1912.17	126.05	1.43	46.59
	0.735	0.1128	813.87	181.82	0.49	42.24
	0.553	0.0013	2276.67	88.90	1.05	37.90
DMSO	0.415	0.0789	823.74	161.75	1.33	31.48
	0.312	0.1406	607.48	171.92	0.11	26.95
	0.234	0.0030	1736.27	70.74	0.89	24.56
	0.172	0.2490	249.14	203.96	0.59	19.35
	0	0.0299	964.27	68.29	0.20	13.36
	0.861	0.0030	1950.82	122.99	0.68	45.93
	0.751	0.0362	1136.57	156.53	0.76	40.40
	0.581	0.0915	842.54	169.93	1.21	36.27
DMA	0.445	0.1719	640.66	176.45	1.21	30.49
DMA	0.343	0.1668	547.88	176.39	0.83	26.05
	0.257	0.0282	1092.06	92.89	0.60	18.87
	0.187	0.2136	311.70	175.77	0.32	14.68
	0	0.0504	632.35	82.11	0.14	9.89
	0.842	0.0380	1227.37	155.39	0.49	42.96
	0.723	0.0328	1139.00	157.09	1.37	40.80
	0.530	0.0786	961.50	150.06	0.20	31.35
DME	0.407	0.0896	784.93	158.19	0.28	28.54
DMF	0.299	0.0734	850.06	133.74	0.25	22.63
	0.224	0.1019	594.21	129.85	0.97	15.12
	0.160	0.0834	616.26	112.00	0.36	12.89
	0	0.0996	328.56	142.97	0.28	9.78

TABLE 6: The parameters for (8), the average absolute relative deviations, and the energy barriers at 303.15 K.

Table 7: Experimental viscosities of [C8mim][OAc] with organic solvent of DMSO at atmospheric pressure (0.0967  $\pm$  0.0020) MPa from 303.15 K to 348.15 K.

			х	[C <sub>8</sub> mim][OA	[c] + (1-x) DN	ISO, η/mPa·s			
T/K					x				
	1.000	0.842	0.735	0.553	0.415	0.312	0.234	0.172	0.000
303.15	304.48	158.20	92.56	53.50	26.70	14.40	5.31	3.08	1.81
308.15	222.12	119.37	70.25	42.71	21.92	12.12	4.61	2.70	1.67
313.15	165.60	91.58	55.59	33.99	18.14	10.38	3.91	2.44	1.54
318.15	126.70	66.17	44.57	27.14	15.36	8.95	3.45	2.21	1.42
323.15	99.22	52.28	35.52	21.81	13.12	7.82	2.93	2.03	1.31
328.15	77.50	41.75	29.46	17.45	11.27	6.85	2.55	1.86	1.22
333.15	62.51	33.13	24.40	14.38	9.22	6.10	2.24	1.69	1.14
338.15	50.11	26.94	20.52	12.11	8.27	5.42	2.02	1.59	1.07
343.15	41.05	22.15	17.57	10.02	7.42	4.88	1.78	1.48	1.00
348.15	33.87	18.29	14.91	8.38	6.79	4.41	1.61	1.41	0.94

The relative combined standard uncertainty  $(u_{c,r})$  is  $u_{c,r}(\eta) = 0.06$ .

parameters of  $C_0$ ,  $C_1$ ,  $C_2$ , and  $C_3$  are fitted by the experimental densities.

Table S4 lists the fit parameters for (7). The calculated density data of the three mixtures are compared with the experiment values. The AARD is 0.11% for IL with DMSO, 0.20% for IL with DMA, and 0.22% for IL with DMF.

3.2. Experimental Viscosity Data and the Effects of Solvents on IL Viscosity. In general, the viscosity can be correlated by the Vogel-Fulcher-Tammann (VFT) equation [46]:

$$\eta = \eta_0 \exp\left[\frac{B}{(T - T_0)}\right],\tag{8}$$

where  $\eta$  (mPa·s) is the viscosity; the parameters of  $\eta_0$  (mPa·s), B (K), and  $T_0$  (K) are fitted using the experiment data; T (K) is the measurement temperature.

Table 6 gives the fit parameters for (8) and AARD. In Figures S7–S9, AARD is 1.96% for DMSO [14, 24, 25], 1.49% for DMA [28, 30, 31, 34], and 1.28% for DMF [14, 20, 38, 47]. It is observed that the deviations between the literature values and the experiment data are desirable. Therefore, the experimental viscosity data of IL with organic solvents are determined and given in Tables 7–9.

Figure 6 and Figures S10–S12 give the experimental viscosity data as a function of IL mole fraction. The viscosity of  $[C_8 mim][OAc]$  is 304.48 mPa s at 303.15 K, while the value decreases to 33.87 mPa s at 348.15 K. The IL viscosity is

Table 8: Experimental viscosities of [C8mim][OAc] with organic solvent of DMA at atmospheric pressure (0.0967  $\pm$  0.0020) MPa from 303.15 K to 348.15 K.

			;	$x [C_8 mim][OA]$	Ac] + (1-x) DI	MA, η/mPa·s			_
T/K					x				
	1.000	0.861	0.751	0.581	0.445	0.343	0.257	0.187	0.000
303.15	304.48	150.44	84.15	51.38	26.85	12.55	5.05	2.46	0.88
308.15	222.12	113.66	65.89	40.02	22.38	10.63	4.56	2.26	0.83
313.15	165.60	85.15	50.60	32.79	18.96	9.28	3.98	2.07	0.78
318.15	126.70	66.05	41.17	27.01	15.84	7.94	3.57	1.91	0.74
323.15	99.22	50.67	33.31	22.98	13.31	6.94	3.22	1.77	0.69
328.15	77.50	39.94	27.56	18.85	11.52	6.18	2.92	1.65	0.66
333.15	62.51	32.10	22.65	15.80	10.14	5.45	2.66	1.54	0.62
338.15	50.11	26.22	18.75	13.82	8.99	4.82	2.45	1.45	0.60
343.15	41.05	20.93	15.81	11.42	8.10	4.49	2.20	1.38	0.57
348.15	33.87	17.45	13.78	10.44	7.40	4.13	2.02	1.31	0.54

The relative combined standard uncertainty  $(u_{c,r})$  is  $u_{c,r}(\eta) = 0.06$ .

Table 9: Experimental viscosities of [C8mim][OAc] with organic solvent of DMF at atmospheric pressure (0.0967  $\pm$  0.0020) MPa from 303.15 K to 348.15 K.

			:	x [C <sub>8</sub> mim][O <sub>4</sub>	Ac] + (1-x) Di	MF, η/mPa·s			
T/K					x				
	1.000	0.842	0.723	0.530	0.407	0.299	0.224	0.160	0.000
303.15	304.48	154.05	79.62	42.01	20.19	11.07	3.15	2.09	0.78
308.15	222.12	117.48	62.38	34.41	16.69	9.60	2.85	1.93	0.72
313.15	165.60	90.95	48.63	28.49	14.26	8.42	2.59	1.79	0.68
318.15	126.70	71.44	38.34	23.99	12.15	7.35	2.38	1.66	0.65
323.15	99.22	57.67	31.45	20.36	10.46	6.49	2.18	1.54	0.62
328.15	77.50	46.09	25.35	17.43	9.07	5.83	2.08	1.45	0.59
333.15	62.51	37.80	20.38	15.00	7.95	5.23	1.93	1.34	0.56
338.15	50.11	31.33	17.83	13.02	7.05	4.68	1.74	1.27	0.54
343.15	41.05	26.66	14.99	11.47	6.24	4.26	1.63	1.20	0.51
348.15	33.87	21.93	13.38	10.03	5.56	3.87	1.56	1.14	0.49

The relative combined standard uncertainty  $(u_{c,r})$  is  $u_{c,r}(\eta) = 0.06$ .

TABLE 10: The parameters for the hard-sphere theory.

IL/solvent	$a \times 10^5$	$b \times 10^3$	$R_{\eta}$	AARD	MD
[C8mim][OAc]	19.001	10.472	1.204	0.38	0.71
DMSO	3.644	4.340	1.549	0.11	0.28
DMA	5.271	3.266	1.263	0.16	0.32
DMF	2.333	6.984	2.604	0.27	0.72

Table 11: The parameters k12 of the hard-sphere theory for the binary mixture.

Mixture	$k_{12}$	AARD
IL-DMSO	0 0.502	20.6 19.7
IL-DMA	0 0.942	21.2 15.6
IL-DMF	0 1.109	25.9 16.8

clearly affected by the temperature. The hydrogen bond is proposed as the main factor for governing the interaction of IL viscosity. The high temperature will dramatically weaken the hydrogen-bond intensity and IL viscosity decreases accordingly [40].

Regarding the binary mixture, for instance, at 303.15 K, the viscosity of [C<sub>8</sub>mim][OAc] with DMSO drops from 304.48 mPa·s to 158.20 mPa·s when the mole fraction of solvent increases from 0 to 0.158 (the mass fraction is 0.05); the viscosity decreases to 92.56 mPa·s when the mole fraction of solvent increases to 0.265 (the mass fraction is 0.10); the viscosity decreases to 53.50 mPa·s when the mole fraction of solvent increases to 0.447 (the mass fraction is 0.20). It is concluded that a small quantity of the solvent has a great effect on lowering the IL high viscosity. The similar results are observed for IL with DMA or DMF. As shown in Figure 6, there are no significant discrepancies for the effects of the studied solvents on lowering the ionic liquid viscosity.

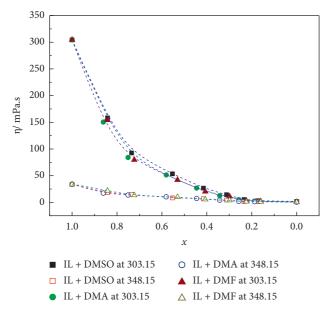


FIGURE 6: Experimental viscosity data of [C8mim][OAc] with solvents as a function of ionic liquid mole fraction at normal pressure.

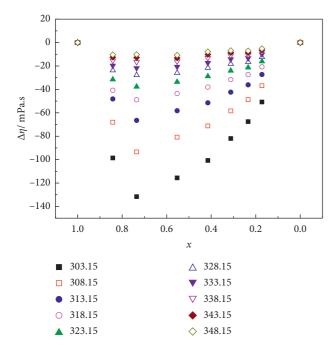


FIGURE 7: The viscosity deviation for the binary mixture of [C8mim][OAc] with solvent of DMSO as a function of ionic liquid mole fraction at atmospheric pressure.

According to the experimental data, the energy barrier  $E_{\eta}$  is introduced and it is a potential field that can be used to either localize or regulate the transfer of charged particles, for example, electrons [48]. Therefore, the higher the energy

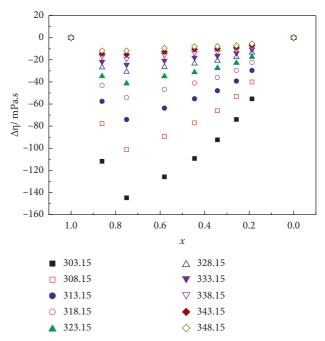


FIGURE 8: The viscosity deviation for the binary mixture of [C8mim][OAc] with solvent of DMA as a function of ionic liquid mole fraction at atmospheric pressure.

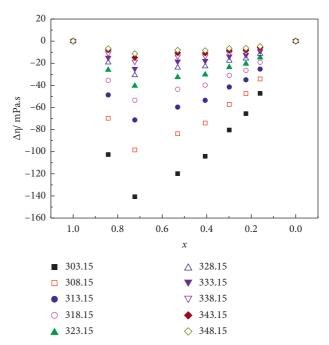


FIGURE 9: The viscosity deviation for the binary mixture of [C8mim][OAc] with solvent of DMF as a function of ionic liquid mole fraction at atmospheric pressure.

barrier value is, the more difficult it is for the ions to move or be regulated, and the energy barrier  $E_{\eta}$  is calculated by [46]

$$E_{\eta} = R \times \frac{\partial \ln \eta}{\partial (1/T)} = R \left( \frac{B}{\left( \left( T_0^2 / T^2 \right) - \left( 2T_0 / T \right) + 1 \right) \right)}, \tag{9}$$

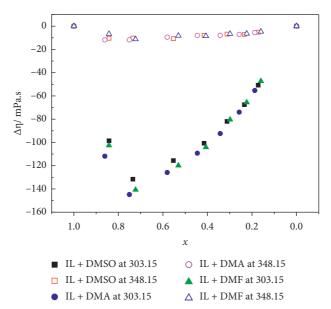


FIGURE 10: The viscosity deviation for the binary mixture of [C8mim][OAc] with solvents as a function of ionic liquid mole fraction at atmospheric pressure.

where R is the ideal gas constant and it is approximately 8.314 JK<sup>-1</sup> mol<sup>-1</sup>;  $\eta$  (mPa·s) is the viscosity; B (K) and  $T_0$  (K) are obtained from (8).

Table 6 gives the energy barrier at 303.15 K. The value of the energy barrier decreases with the increase of the solvent content in the binary mixture that is consistent with the phenomenon of the decrease for the binary mixture viscosity when the solvent content in the mixture increases.

Based on the experiment data, the viscosity deviation,  $\Delta \eta$ , is calculated to analyze the effect of the solvent on IL viscosity according to the following equation:

$$\Delta \eta = \eta - \sum x_i \eta_i, \tag{10}$$

where  $\eta$  and x are the viscosity and mole fraction, respectively. The subscript of "i" means the property of pure substance i. The relative combined standard uncertainty of the viscosity deviation is 0.14.

Figures 7–10 give the viscosity deviation as a function of ionic liquid mole fraction at atmospheric pressure. In the studied temperatures, the deviations are all negative and the absolute values increase with the decrease of temperature because IL viscosity at high temperature is at the same order of magnitude with that of the solvent and, then, the viscosity deviations at high temperature become smaller. The curves are asymmetric and the minimum values are detected at the concentrated area.

In the previous studies, spectra of IL presented that a delocalization of the electrons in ionic acetate anion and the interactions with IL cation were observed; moreover, the comparatively weak C–H stretching was detected, whereas a broad band corresponding to the intermolecular O–H vibrations caused by the H-bond was determined [49]. It is therefore reasonable to infer that there are possible H-bond intermolecular interactions between the O–H vibrations in IL cation with S=O in DMSO, C=O in DMA, and DMF.

Furthermore, the hard-sphere theory is used to study the binary viscosity. The reduced viscosity of rough hard-sphere  $\eta_{\text{RHS}}^*$  is related to the reduced viscosity of smooth hard-sphere  $\eta_{\text{SHS}}^*$  through a proportionally constant  $R_{\eta}$  [50–52]:

$$\eta_{\rm RHS}^* = R_\eta \eta_{\rm SHS}^*. \tag{11}$$

The reduced viscosity of rough hard sphere is calculated as follows:

$$\eta_{\text{RHS}}^* = 6.035 \times 10^8 \times (MRT)^{-0.5} \eta V^{(2/3)},$$
(12)

where M (kg·mol<sup>-1</sup>) is the molecular weight, R is the universal gas constant and it is 8.3141 J mol<sup>-1</sup> K<sup>-1</sup>, T (K) is the temperature,  $\eta$  (Pa·s) is the viscosity, and V (m<sup>3</sup>·mol<sup>-1</sup>) is the molar volume.

The reduced viscosity of smooth hard sphere is expressed as follows:

$$\log_{10}(\eta_{\text{SHS}}^*) = \log_{10}\left(\frac{\eta_{\text{RHS}}^*}{R_{\eta}}\right) = \sum_{i=0}^{7} a_{\eta i} \left(\frac{V}{V_0}\right)^{-i}.$$
 (13)

 $V/V_0$  is defined as the reduced molar volume  $V_r$  and  $V_0$  (m<sup>3</sup>·mol<sup>-1</sup>) is a characteristic molar volume. Ciotta et al. extended the expression to high-viscosity hydrocarbons and the coefficients  $a_{\eta i}$  are 0, 5.14262, -35.5878, 192.05015, -573.37246, 957.41955, -833.36825, and 299.40932 [53]. The characteristic molar volume  $V_0$  is proposed as follows [52]:

$$V_0 = a + b/T, \tag{14}$$

where the parameters of a, b, and  $R_{\eta}$  are fitted by the viscosity data in this work. Table 10 gives the fit parameters, AARD, and the maximum absolute relative deviation (MD).

For the binary mixture, the mixing rule for the binary mixture of IL with water proposed by Teja et al. is employed for IL with solvents [52]:

$$\log(\eta_{\text{mix}}) = x_1 \log(\eta_1) + x_2 \log(\eta_2) + 2x_1 x_2 k_{12} \left(\frac{M_1 M_2}{M_{\text{mix}}}\right)^{(1/2)},$$
(15)

where  $k_{12}$  is a binary interaction parameter. Table 11 presents the parameter  $k_{12}$  and AARD. The deviations are large for the binary mixture. The deviations are larger than 20% without the adjustable parameter of  $k_{12}$ , but they are reduced to less than 20% when  $k_{12}$  is used. Teja et al. reported the deviations in the range of 7.10%–16.37% using the mixing rule for the viscosity prediction of water with IL [52]. The addition of solvent significantly lowers the IL viscosity, and the above analysis indicates that H-bond intermolecular interactions occur between ionic liquid and solvents; therefore, the mixing rules are required for further study and improvement.

#### 4. Conclusions

In this work, the thermophysical properties of the densities and viscosities for [C<sub>8</sub>mim][OAc] with the organic solvents (i.e., DMSO, DMA, and DMF) were studied at normal pressure in the temperature ranges from 303.15 K to 348.15 K. The excess molar volume was calculated and the values are negative under the studied temperatures, indicating that the volumes of binary mixtures become smaller than the ideal ones and there should be interactions between the IL and the solvents. Regarding the viscosity, a small proportion of solvent would lower ionic liquid viscosity. Among the three solvents, there are no apparent distinctions on lowering IL viscosity. Summarily, the small deviations for the density from the ideality are observed, while larger deviations for the viscosity are detected. The rough hardsphere model works well to reproduce the viscosity behavior of the pure substances, while the mixing rules for the binary mixture merit further study.

#### **Data Availability**

The data are available in the Supplementary Material: the parameters fitted in this work for (2) and (5); the deviation for solvents between the literature density/viscosity and the experimental data in this work; experimental density/viscosity data of ionic liquid with solvents as a function of the temperature or IL mole fraction.

## **Conflicts of Interest**

The authors declare that they have no conflicts of interest.

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### **Supplementary Materials**

The following are provided: chemical structure and physical properties of IL; the parameters fitted in this work for (2),

(5), and (7); the deviation for solvents between the literature density/viscosity and the experimental data in this work; experimental density/viscosity data of ionic liquid with solvents as a function of the temperature or IL mole fraction. (Supplementary Materials)

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